

## Evidence for New Isotopes of Element 107: $^{266}\text{Bh}$ and $^{267}\text{Bh}$

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New neutron rich isotopes,  $^{267}\text{Bh}$  and  $^{266}\text{Bh}$ , were produced in bombardments of a  $^{249}\text{Bk}$  target with 117-MeV and 123-MeV  $^{22}\text{Ne}$  ions at the Lawrence Berkeley National Laboratory 88-Inch Cyclotron. Identification was made by observation of correlated  $\alpha$ -particle decays between the Bh isotopes and their Db and Lr daughters using a rotating wheel system.  $^{267}\text{Bh}$  was produced with a cross-section of  $\approx 70$  pb and decays with a  $17^{+14}_{-6}$  s half-life by emission of  $\alpha$  particles with an average energy of  $8.83 \pm 0.03$  MeV. One atom of  $^{266}\text{Bh}$  was observed, decaying within one second by emission of a 9.29-MeV  $\alpha$  particle.

Element 107, (bohrium, Bh) [1], was first identified as the isotope  $^{262}\text{Bh}$  produced in the  $^{209}\text{Bi}(^{54}\text{Cr},n)$  reaction [2] in 1981 by Münzenberg *et al.* using the velocity filter SHIP at GSI Darmstadt. Previous to our experiment, only the decay properties of isotopes  $^{261}\text{Bh}$  ( $T_{1/2}=11.8$  ms;  $E_{\alpha}= 10.40, 10.10, 10.03$  MeV),  $^{262}\text{Bh}$  ( $T_{1/2}=102$  ms;  $E_{\alpha}= 10.06, 9.91, 9.74$  MeV),

$^{262}\text{Bh}^m$  ( $T_{1/2}=8$  ms;  $E_\alpha=10.37, 10.24$ ), and  $^{264}\text{Bh}$  ( $T_{1/2}=440$  ms;  $E_\alpha=9.62, 9.48$  MeV) were well known [3]. This experiment was undertaken to try to produce and identify the new neutron-rich isotopes of bohrium,  $^{266}\text{Bh}$  and  $^{267}\text{Bh}$ . These isotopes were predicted to have significantly longer half-lives than previously known bohrium isotopes [4][5], possibly long enough to enable the first studies of bohrium chemical properties in subsequent experiments.

Based on predicted Q-values for electron capture and a decay [5],  $^{266}\text{Bh}$  and  $^{267}\text{Bh}$  should decay predominantly by  $\alpha$ -emission and possibly by spontaneous fission (SF). The  $\alpha$ -particle energies and half-lives for these isotopes are expected to be in the range of 8.7–9.3 MeV [5] and 1-20 seconds [4]. The previously reported decay characteristics [3] of their Db and Lr daughter nuclei, summarized in Fig. 1, are:  $^{262}\text{Db}$  ( $T_{1/2}=34$  s;  $E_\alpha=8.45, 8.53, 8.67$  MeV),  $^{263}\text{Db}$  ( $T_{1/2}=27$  s;  $E_\alpha=8.35$  MeV),  $^{258}\text{Lr}$  ( $T_{1/2}=3.9$  s;  $E_\alpha=8.60, 8.62, 8.57, 8.65$  MeV), and  $^{259}\text{Lr}$  ( $T_{1/2}=6.34$  s;  $E_\alpha=8.45$  MeV). The *Table of Isotopes* [3] lists an  $\alpha$ -particle energy of 8.35 MeV for  $^{263}\text{Db}$ , from Kratz *et al.* [6], but other measurements [7] indicate that  $^{263}\text{Db}$  might also decay by emission of 8.41-MeV  $\alpha$  particles.

In our experiment, the nuclides  $^{267}\text{Bh}$  and  $^{266}\text{Bh}$  were produced via the  $^{249}\text{Bk}(^{22}\text{Ne}, 4n)$  and  $^{249}\text{Bk}(^{22}\text{Ne}, 5n)$  reactions. The Lawrence Berkeley National Laboratory 88-Inch Cyclotron provided beams of 148-MeV, 2-euA  $^{22}\text{Ne}^{6+}$  and 153-MeV, 2-euA  $^{22}\text{Ne}^{6+}$ . The target system has been described previously [8]. The beam enters through a 2.73-mg/cm<sup>2</sup> beryllium vacuum window, then passes through 0.5 mg/cm<sup>2</sup> of nitrogen cooling gas and a 2.73-mg/cm<sup>2</sup> beryllium target backing, before passing through the target material, 0.81 mg/cm<sup>2</sup> of  $^{249}\text{Bk}$  as the oxide prepared by the molecular plating technique [9][10]. The  $^{249}\text{Cf}$  daughter ( $T_{1/2}=351$  yr) was separated from the  $^{249}\text{Bk}$  target material ( $T_{1/2}=325$  d) five days prior to the beginning of the experiment with less than 0.5% (atom)  $^{249}\text{Cf}$  present after separation. The 148-MeV and

153-MeV  $^{22}\text{Ne}^{6+}$  beams resulted in projectile energies in the  $^{249}\text{Bk}$  target of 116 - 118 MeV and 122 - 124 MeV, respectively. Products of the nuclear reactions recoiled out of the target and through the  $50\text{-}\mu\text{g}/\text{cm}^2$  Al cover foil. A recoil chamber, located directly behind the target, was continuously swept with He gas (2.5 l/min, 1.2 atm) containing KCl aerosols to collect the reaction products. These products were then transported through a Teflon capillary (1.4 mm-diameter, 7 m-long) to the Merry-Go-round (MG) rotating wheel system [11][12] with an approximate transport time of 0.6 s including retention time in the recoil chamber with a transport efficiency of  $38\pm 4\%$ . The transport efficiency was determined by comparing the spontaneous fission activity from Fm-Md transfer products detected at the MG with that measured in a catcher-foil placed directly behind the target.

The MG detection system consists of a 20-inch-diameter fiberglass rotating wheel, which is rotated between six pairs of passivated, ion-implanted planar silicon (PIPS) detectors. The data from the second top detector was excluded from the analysis due to a malfunction indicated by an excessive amount of noise. For this experiment, a parent-daughter stepping mode was used to provide detection of  $\alpha$ - $\alpha$  correlations with a greatly reduced background [13]. The reaction products are deposited on 40 polypropylene films ( $\sim 40\text{ }\mu\text{g}/\text{cm}^2$ ) placed in every other position around the periphery of the 80-position rotating collection wheel. Every ten seconds during parent-search mode, the wheel is double-stepped between the six pairs of  $\alpha$ -particle detectors until a possible parent decay is detected in a bottom detector. If an  $\alpha$  particle is detected in the bottom detector within an energy window that is expected for  $^{266}\text{Bh}$  or  $^{267}\text{Bh}$  (between 8.6 and 10.5 MeV), it is assumed that the daughter dubnium nucleus,  $^{263}\text{Db}$ , has recoiled out of the KCl sample and into the top detector. The probability of the daughter nucleus recoiling into the top detector was previously measured at about 65%. When a possible parent decay event is detected,

a 60-s daughter-search mode is initiated by single stepping the wheel to move an empty position between the detectors in order to detect the daughter or granddaughter  $\alpha$ -decay in the absence of the activity on the collection foil. At the end of the daughter mode interval, the wheel is single stepped again and parent-search mode is resumed.

The  $\alpha$ -particle energy resolution above the wheel was  $\sim 40$  keV, while the energy resolution of the detectors below was  $\sim 100$  keV due to energy degradation in the polypropylene foils. The energy calibration was performed on-line using known  $\alpha$  decay energies of  $^{212}\text{Ra}$  ( $E_\alpha = 6.901$  MeV) and  $^{212}\text{Po}^m$  ( $E_\alpha = 11.650$  MeV). The representative parent-mode  $\alpha$ -spectrum shown in Fig. 2 illustrates the very low overall  $\alpha$ -counting rate and the relatively small contribution from Ra, Rn, and Po isotopes. Fm and No isotopes formed by low-yield transfer reactions are also visible. The total event rate seen by the detector array was about 5 events per second. The first detector pair saw 75% of the event rate, with the remaining 25% distributed equally among the other five detectors. The top and bottom detectors of an individual detector pair were exposed to approximately the same rate.

An off-line computer search was made for  $\alpha$ - $\alpha$  correlations between Bh events ( $8.6 < E_\alpha(\text{MeV}) < 10.5$ ) in parent-mode followed by daughter  $\alpha$ -events ( $8.2 < E_\alpha(\text{MeV}) < 8.7$ ) detected in the same detector pair during the ensuing daughter mode search. Five atoms of  $^{267}\text{Bh}$ ,  $E_\alpha$  ranging from 8.73 to 8.87 MeV, and one atom of  $^{266}\text{Bh}$  with an  $E_\alpha$  of 9.29 MeV, were identified during the experiment (See Table I). Possible summing with conversion electrons from population of levels above the ground state and energy degradation in the polypropylene foil make it impossible to determine conclusively if more than one  $^{267}\text{Bh}$   $\alpha$ -group is present. The average  $\alpha$ -energy is  $8.83 \pm 0.03$  MeV. The measured  $^{267}\text{Bh}$  half-life, using the maximum likelihood technique (MLDS) [14] allowing all parameters vary with a single component fit, and

taking into account the timing constraints of the measurement, is  $17^{+14}_{-6}$  s. The five a-events attributed to the a-decay of  $^{267}\text{Bh}$  daughter nuclei are consistent with  $^{263}\text{Db}$  and  $^{259}\text{Lr}$ . Assuming a-decay is the dominant decay mode, the  $^{249}\text{Bk}(^{22}\text{Ne},4n)^{267}\text{Bh}$  cross section is  $58^{+33}_{-15}$  pb at 116 to 118 MeV and is  $96^{+55}_{-25}$  pb at 122 to 124 MeV. Using the SPIT code [15], we calculated that  $\sim 32$  pb at 117 MeV is the maximum cross-section for the  $^{249}\text{Bk}(^{22}\text{Ne},4n)^{267}\text{Bh}$  reaction and it is  $\sim 14$  pb 123 MeV.

Of the 609 events during the experiment that initiated daughter mode, about half of them occurred in the first detector with the balance fairly evenly distributed among the remaining five detectors. About 13% of the time during the experiment was spent in daughter mode. We observed five cases during the entire experiment where a-events in daughter mode with  $8.2 < E_a(\text{MeV}) < 8.7$  were observed in detector pairs different from that in which the initiating parent event was observed. Assuming that these random daughter events are evenly distributed among the detectors, we estimate that approximately one of the five  $^{267}\text{Bh}$  a-a correlations reported is due to a random correlation of unrelated a-decays. Based on this random daughter rate, the expected number of random a-a-a triple correlations is 0.0016.

During the entire experiment, there was only one instance where a potential parent event at 9.29 MeV was followed by two a particles with  $8.2 < E_a(\text{MeV}) < 8.7$  in the daughter mode. The details of this event number 6, are listed at the bottom of Table I. The daughter-mode energies and lifetimes are consistent with those expected for  $^{262}\text{Db}$  and  $^{258}\text{Lr}$ . On this basis, we assign the 9.29-MeV event to the decay of  $^{266}\text{Bh}$  produced in the  $^{249}\text{Bk}(^{22}\text{Ne},5n)$  reaction. This triple correlation occurred during the higher energy bombardment, supporting assignment of the  $5n$ -exit channel.

Since the wheel stepping time in this experiment was 10 s to optimize the search for isotopes in the 10-30 s range, the  $^{266}\text{Bh}$  production cross section obtained from this measurement is strongly dependent on the assumed  $^{267}\text{Bh}$  half-life. According to  $\alpha$ -decay systematics [4] the unhindered half-life for 9.29-MeV  $^{266}\text{Bh}$  decay should be  $\sim 0.5$  s. An  $\alpha$ -hindrance factor between 2 and 20 would correspond to a half-life of 1-10 s. A half-life in this range would indicate a cross section between 250 and 25 pb, taking into account the experimental conditions. The cross section calculated with SPIT was 3 pb at 122 to 124 MeV.

The new nuclides  $^{266}\text{Bh}$  and  $^{267}\text{Bh}$  have been observed to decay via  $\alpha$  emission.  $^{267}\text{Bh}$  has a half-life of  $17^{+14}_{-6}$  s, and emits  $\alpha$ -particles with an average energy of  $8.83 \pm 0.03$  MeV. One event with an  $\alpha$ -particle energy of 9.29 MeV and an estimated half-life of 1-10 s was attributed to  $^{266}\text{Bh}$  based on the observed triple  $\alpha$ -correlation. We were not able to determine the fission decay properties of either isotope due to fission contamination that is attributed to  $^{256}\text{Fm}$ ; about 12 fissions per hour were measured in coincidence in the top and bottom of each detector pair. As shown in Table 2, the measured cross sections of 25-250 pb and  $58^{+33}_{-15} / 96^{+55}_{-25}$  pb for  $^{266}\text{Bh}$  and  $^{267}\text{Bh}$ , respectively, are in agreement with the observed trends between  $4n$  and  $5n$  exit channels for analogous reactions [6][12][16][17][18]. Our measured production cross section for  $^{267}\text{Bh}$  is consistent with expectations based on calculations and these systematics. The lifetime of the new isotope  $^{267}\text{Bh}$  is sufficient for studies of the chemical properties of element 107 in either the aqueous or gas phase with fast separation techniques currently in use [19].

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TABLES:

TABLE I. List of correlation between parents events ( $8.6 < E_a(\text{MeV}) < 10.5$ ) and daughter events ( $8.2 < E_a(\text{MeV}) < 8.7$ ). The initiating parent event, each subsequent  $\alpha$ -decay that occurred within the energy window, its isotope assignment, a energy, and relative time are listed for each event.

	Parent	$a_1$	time <sup>c</sup>	Isotope	$a_2$	time <sup>d</sup>	Isotope	$a_3$	time <sup>e</sup>
1 <sup>a</sup>	<sup>267</sup> Bh	8.83 MeV	5.26 s	<sup>263</sup> Db or <sup>259</sup> Lr	8.47 MeV	59.04 s			
2 <sup>a</sup>	<sup>267</sup> Bh	8.87 MeV	24.67 s	<sup>263</sup> Db	8.39 MeV	35.02 s			
3 <sup>a</sup>	<sup>267</sup> Bh	8.87 MeV	45.15 s	<sup>263</sup> Db	8.39 MeV	24.49 s			
4 <sup>b</sup>	<sup>267</sup> Bh	8.73 MeV	2.71 s	<sup>263</sup> Db or <sup>259</sup> Lr	8.46 MeV	51.90 s			
5 <sup>b</sup>	<sup>267</sup> Bh	8.84 MeV	21.83 s	<sup>263</sup> Db	8.36 MeV	26.49 s			
6 <sup>b</sup>	<sup>266</sup> Bh	9.29 MeV	0.87 s	<sup>262</sup> Db	8.54 MeV	27.83 s	<sup>258</sup> Lr	8.74 MeV	0.04 s

<sup>a</sup> 116-118 MeV <sup>22</sup>Ne

<sup>b</sup> 122-124 MeV <sup>22</sup>Ne

<sup>c</sup> time after end of 10-s collection

<sup>d</sup> time after  $a_1$

<sup>e</sup> time after  $a_2$

TABLE II. Reactions analogous to the <sup>22</sup>Ne on <sup>249</sup>Bk reaction showing similar  $4n$  and  $5n$  exit channel systematics.

Reaction	$5n$	$4n$	References
<sup>249</sup> Bk( <sup>22</sup> Ne, $xn$ ) <sup>266,267</sup> Bh	25-250 pb	$58^{+33}_{-15}$ pb, $96^{+55}_{-25}$ pb	this work
<sup>248</sup> Cm( <sup>22</sup> Ne, $xn$ ) <sup>265,266</sup> Sg	240 pb	80 pb	[16,17]
<sup>249</sup> Bk( <sup>18</sup> O, $xn$ ) <sup>263,262</sup> Db	6 nb	2 nb	[6]
<sup>244</sup> Pu( <sup>22</sup> Ne, $xn$ ) <sup>262,261</sup> Rf	3 nb	0.7 nb	[12,18]





Fig. 1

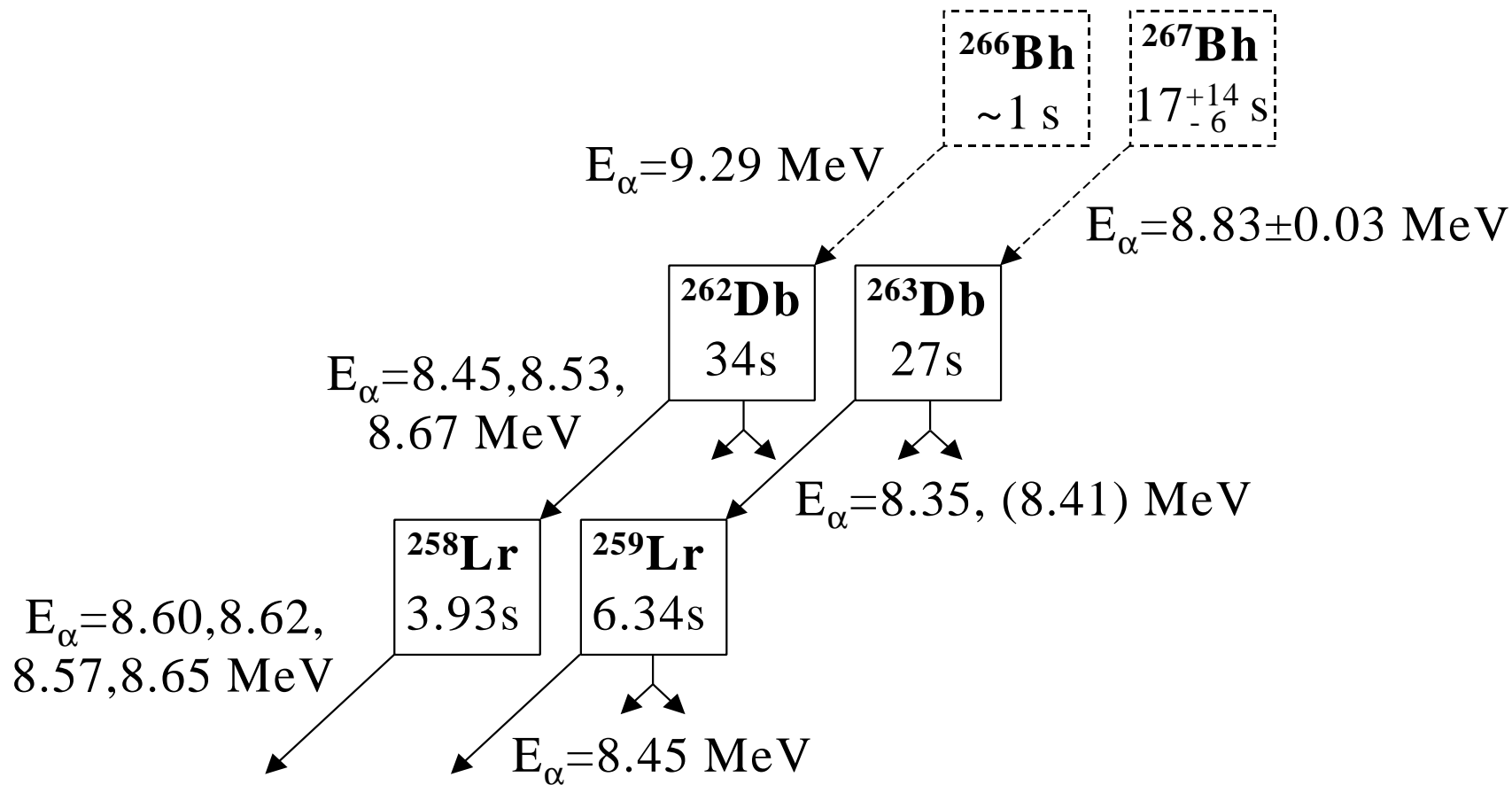
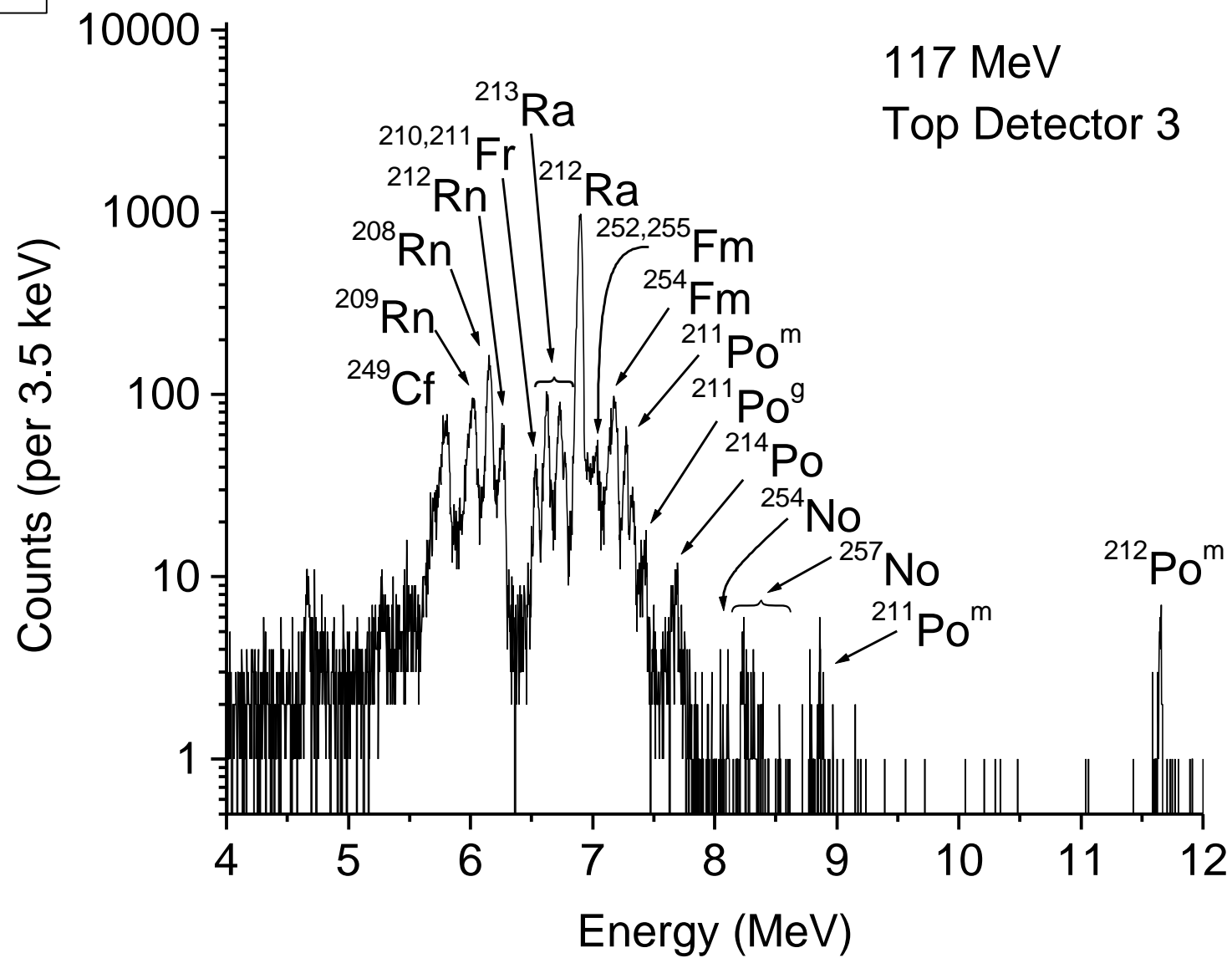


Fig. 2



CAPTIONS:

FIG. 1. Partial decay chain of  $^{266}\text{Bh}$  and  $^{267}\text{Bh}$  [3,7]. Decay properties of  $^{266}\text{Bh}$  and  $^{267}\text{Bh}$  in the dashed boxes are as measured during this experiment.

FIG. 2. The  $\alpha$ -particle spectrum measured over the entire 62-h experiment in detector pair three top, of products of the reaction of 117-MeV  $^{22}\text{Ne}$  with  $^{249}\text{Bk}$ .